# FILING BY "EXPRESS MAIL" UNDER 37 CFR 1.10

A substitute specification.

A change of power of attorney and/or address letter.

Other items or information: (See attached Form PCT/ISA/210)

14. 🗆

15. 🗆

16. 🗹

(REV 10-95)		Department of Commerce Faters and Trademark Office	HF/5-22102/A/PCT							
TRANSMITTAL LETTER TO		THE UNITED STATES	U.S. APPLICATION NO. (If known, see 37 CFR 1.5)							
	DESIGNATED/ELECTED O	•0/0000-0								
	CONCERNING A FILING UN	10/089852								
INTERNATIONAL APPLICATION NO.		INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED							
PCT/EP 00/09396		September 26, 2000	October 5, 1999							
TITLE OF INVENTION										
IMPROVED FABRIC SOFTENER COMPOSITIONS  APPLICANT(S) FOR DO/EO/US										
Petr Kvita, Peter Otto, Mario Dubini, Harald Chrobaczek, Michael Geubtner, Ralf Goretzki, Barbara Weber and										
Emmanuel Martin										
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:										
1. 🗹	This is a <b>FIRST</b> submission of items cor	ncerning a filing under 35 U.S.C. 371.								
2. 🗆	This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.									
3.	This express request to begin national examination procedures (35 U.S.C. 371(f) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).									
4. 🗹	A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority									
5. ☑	date.  A copy of the International Application a	is filed (35 U.S.C. 371(c)(2))								
J. E	<ul> <li>✓ A copy of the International Application as filed (35 U.S.C. 371(c)(2))</li> <li>a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).</li> </ul>									
	b.   has been transmitted by the In	ternational Bureau. (See attached For	m PCT/IB/308)							
6. □		ion was filed in the United States Receivation into English 35 U.S.C. 371(c)(2)).	Ving Office (RO/OS).							
0. □ 7. ☑										
	<ul> <li>a.    ☐ are transmitted herewith (required only if not transmitted by the International Bureau).</li> </ul>									
	<ul><li>b.  have been transmitted by the</li><li>c.  have not been made; however</li></ul>	International Bureau. ·, the time limit for making such amendm	nents has NOT expired							
	d. ✓ have not been made and will r		ionio nao von oxpirodi							
8. 🗆	A translation of the amendments to the	claims under PCT Article 19 (35 U.S.C.	371 (c)(3)).							
9. 🗆										
10. 🗆	A translation of the annexes to the inter	Hational Preliminary Examination Nepol	t under FOT Article 30 (33 0.3.0. 37 1(5)(3)).							
items 1	1. to 16. below concern document(s) o	r information included.								
11. 🗆	An Information Disclosure Statement ur	nder 37 CFR 1.97 and 1.98.								
12. 🗆	An assignment document for recording.	A separate cover sheet in compliance	with 37 CFR 3.28 and 3.31 is included.							
13. 🗹	A FIRST preliminary amendment.									
	A SECOND or SUBSEQUENT prelimina	ary amendment.								

						S DOCKET NUMBER 1102/A/PCT				
17.   ✓ The following	fees are submitted:				\$890.00	CALCULATIONS	PTO USE ONLY			
17. ☑ The following fees are submitted: \$890.00 CALCULATIONS PTO USE ONLY  BASIC NATIONAL FEE (37 CFR 1.492(a) (1)-(5)):										
27.10.10 (11.11.10.11.11.11.11.11.11.11.11.11.11.1										
Search Report h										
International preliminary examination fee paid to USPTO (37 CFR 1.482)										
No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)) \$740.00										
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO\$1040.00										
International pre and all claims sa		-								
	\$890.00									
Surcharge of \$130.00 f months from the earlies	30	\$								
CLAIMS	NUMBER FILE		NUMBER EXTRA	RA	TE					
Total claims	20 - 2	20 =	0	´ X \$1		\$				
Independent claims		-3 =	0		4.00	\$				
MULTIPLE DEPENDE	NT CLAIM(S) (if appl				80.00	\$				
	<u> </u>		TAL OF ABOVE C			\$890.00				
	Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28).									
	TOTAL =	\$890.00								
Processing fee of \$130 months from the earlies	\$									
	\$890.00									
Fee for recording the e	nclosed assignment	(37 CFR	TOTAL N. 1,21(h)). The assignm			\$				
accompanied by an ap					+	•				
			TOTAL FI	ES ENCL	OSED =	\$				
		Amount to be:	\$							
						refunded				
		charged	\$890.00							
a. ☑ Please charge my Deposit Account No. 03-1935 in the amount of \$890.00 to cover the above fees. A duplicate copy of this sheet is enclosed.										
b. 🗹 The Commiss	The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to									
Deposit Acco	Deposit Account No. 03-1935. A duplicate copy of this sheet is enclosed.									
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.										
PLEASE ASSOCIATE THE ATTACHED APPLICATION WITH CUSTOMER NUMBER 000324 AND SEND ALL CORRESPONDENCE TO:  JoAnn Villamizar, Ciba Specialty Chemicals Corporation										
toAnn Villamizer Cibe St	Mansfu	Wer .								
JoAnn Villamizar, Ciba Specialty Chemicals Corporation Patent Department SIGNATURE						<i> </i>				
540 White Plains Road										
P.O. Box 2005	eld									
Tarrytown, NY 10591-9005 NAME										
APR () 3 2002 Reg. No. 31,63						5				

### CASE HF/5-22102/A/PCT

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE PCT NATIONAL STAGE APPLICATION OF

**Group Art Unit:** 

PETR KVITA ET AL

Examiner:

INTERNATIONAL APPLICATION NO. PCT/EP 00/09396

FILED: SEPTEMBER 26, 2000

FOR: IMPROVED FABRIC SOFTENER

**COMPOSITIONS** 

U.S. APPLICATION NO: UNASSIGNED

35 USC 371 DATE:

**Assistant Commissioner for Patents** 

Washington, D.C. 20231

# PRELIMINARY AMENDMENT

Sir:

Kindly amend this application as follows prior to calculation of the filing fee and consideration on the merits.

## IN THE CLAIMS

Please cancel claims 1-20.

Please add the following claims.

--21. (new) A method of use of a softener composition for imparting hydrophilicity to textile fibre materials in domestic applications, which comprises treating washed textile fibre materials with a softener composition which comprises:

- A) a fabric softener;
- B) at least one additive selected from the group consisting of
  - a) a polyethylene, or a mixture thereof,
  - b) a fatty acid alkanolamide, or a mixture thereof,
  - c) a polysilicic acid, or a mixture thereof, and
  - d) a polyurethane, or a mixture thereof; and
- C) a dispersed polyorganosiloxane of formula (1)

(1) 
$$R^{1} - S_{1} - O + CH_{3} - CH_{$$

wherein

R<sup>1</sup> is OH, OR<sup>2</sup> or CH<sub>3</sub>,

R<sup>2</sup> is CH<sub>3</sub> or CH<sub>2</sub>CH<sub>3</sub>,

 $R^3$  is  $C_1$ - $C_{20}$ alkoxy,  $CH_3$ ,  $CH_2CHR^4CH_2NHR^5$ , or  $CH_2CHR^4CH_2N(COCH_3)R^5$ ,

or (4) 
$$(CH_2)_3 - N$$
  $NR^8$ 

R<sup>4</sup> is H or CH<sub>3</sub>,

 $R^5$  is H,  $CH_2CH_2NHR^6$ ,  $C(=O)-R^7$  or  $(CH_2)_Z-CH_3$ , z is 0 to 7,

R<sup>6</sup> is H or C(=O)-R<sup>7</sup>, R<sup>7</sup> is CH<sub>3</sub>, CH<sub>2</sub>CH<sub>3</sub> or CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>OH, R<sup>8</sup> is H or CH<sub>3</sub>, and the sum of X and Y is 40 to 4000;

or a dispersed polyorganosiloxane which comprises at least one unit of the formula (5)

(5) 
$$(R^9)_V (R^{10})_W \text{Si-A-B}$$

wherein

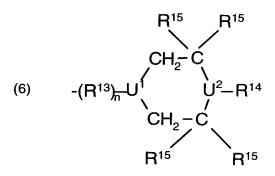
R<sup>9</sup> is CH<sub>3</sub>, CH<sub>3</sub>CH<sub>2</sub> or phenyl,

R<sup>10</sup> is -O-Si or -O-R<sup>9</sup>,

the sum of v and w equals 3, and v does not equal 3,

 $A = -CH_2CH(R^{11})(CH_2)_K,$ 

 $B = -NR^{12}((CH_2)_1-NH)_mR^{12}$  or



n is 0 or 1,

when n is 0, U1 is N, when n is 1, U1 is CH,

I is 2 to 8,

k is 0 to 6,

m is 0 to 3,

R<sup>11</sup> is H or CH<sub>3</sub>,

 $R^{12}$  is H, C(=O)- $R^{16}$ ,  $CH_2(CH_2)_pCH_3$  or

p is 0 to 6,

R<sup>13</sup> is NH, O, OCH<sub>2</sub>CH(OH)CH<sub>2</sub>N(butyl), OOCN(butyl)

R<sup>14</sup> is H, linear or branched C<sub>1</sub>-C<sub>4</sub>alkyl, phenyl or CH<sub>2</sub>CH(OH)CH<sub>3</sub>,

R<sup>15</sup> is H or linear or branched C<sub>1</sub>-C<sub>4</sub>alkyl,

 $R^{16}$  is  $CH_3$ ,  $CH_2CH_3$  or  $(CH_2)_qOH$ ,

q is 1 to 6, and

U<sup>2</sup> is N or CH;

or a dispersed polyorganosiloxane of the formula (8)

(8) 
$$R^{17} - Si - O = \begin{bmatrix} CH_3 \\ | \\ | \\ CH_3 \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ | \\ CH_3 \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix}$$

## wherein

R<sup>3</sup> is as previously defined,

R<sup>17</sup> is OH, OR<sup>18</sup> or CH<sub>3</sub>,

R<sup>18</sup> is CH<sub>3</sub> or CH<sub>2</sub>CH<sub>3</sub>.

 $R^{19}$  is  $R^{20}$ -(EO)<sub>m</sub>-(PO)<sub>n</sub>- $R^{21}$ ,

m is 3 to 25,

n is 0 to 10,

R<sup>20</sup> is the direct bond or CH<sub>2</sub>CH(R<sup>22</sup>)(CH<sub>2</sub>)<sub>p</sub>R<sup>23</sup>,

p is 1 to 4,

 $R^{21}$  is H,  $R^{24}$ ,  $CH_2CH(R^{22})NH_2$  or  $CH(R^{22})CH_2NH_2$ ,

R<sup>22</sup> is H or CH<sub>3</sub>,

R<sup>23</sup> is O or NH.

 $R^{24}$  is linear or branched  $C_1$ - $C_8$ alkyl or  $Si(R^{25})_3$ ,

R<sup>25</sup> is R<sup>24</sup>, OCH<sub>3</sub> or OCH<sub>2</sub>CH<sub>3</sub>,

EO is -CH2CH2O- .

PO is -CH(CH<sub>3</sub>)CH<sub>2</sub>O- or -CH<sub>2</sub>CH(CH<sub>3</sub>)O- and the sum of X<sub>1</sub>,Y<sub>1</sub> and S is 20 to 1500;

or a dispersed polyorganosiloxane of the formula (9)

#### wherein

R<sup>26</sup> is linear or branched C<sub>1</sub>-C<sub>20</sub>alkoxy or CH<sub>2</sub>CH(R<sup>4</sup>)R<sup>29</sup>,

R4 is as previously defined,

R<sup>29</sup> is linear or branched C<sub>1</sub>-C<sub>20</sub>alkyl,

 $R^{27}$  is aryl, aryl substituted by linear or branched  $C_1$ - $C_{10}$ alkyl, linear or branched  $C_1$ - $C_{20}$ alkyl substituted by aryl or aryl substituted by linear or branched  $C_1$ - $C_{10}$ alkyl,  $R^{28}$  is

(10) 
$$(CH_2)_3$$
— $O$ — $CH_2$ — $CH_2$ — $CH_2$ , and

the sum of  $X^2$ ,  $X^3$ ,  $X^4$  and  $Y^2$  is 20 to 1500, wherein  $X^3$ ,  $X^4$  and  $Y^2$  may be independently of each other 0;

or a mixture thereof.

22. (new) A method of use according to claim 21 wherein the polyorganosiloxane is of formula (1):

(1) 
$$R^{1} - S_{1} - O = \begin{bmatrix} CH_{3} & CH_{3} & CH_{3} & CH_{3} \\ S_{1} - O & S_{1} - O \end{bmatrix} = \begin{bmatrix} CH_{3} & CH_{3} & CH_{3} & CH_{3} \\ S_{1} & CH_{3} & CH_{3} & CH_{3} \end{bmatrix}$$

#### wherein

R1 is OH, OR2 or CH3

R<sup>2</sup> is CH<sub>3</sub> or CH<sub>2</sub>CH<sub>3</sub>

R<sup>3</sup> is C<sub>1</sub>-C<sub>20</sub>alkoxy, CH<sub>3</sub>, CH<sub>2</sub>CHR<sup>4</sup>CH<sub>2</sub>NHR<sup>5</sup>, or

(2) 
$$(CH_2)_3O$$
 NR<sup>8</sup> or (3)  $(CH_2)_3NH$  CH

 $R^4$  is H or  $CH_3$ ,  $R^5$  is H,  $CH_2CH_2NHR^6$ ,  $C(=O)-R^7$ ,  $R^6$  is H or  $C(=O)-R^7$ ,

 $\mbox{R}^{7}$  is  $\mbox{CH}_{3},$   $\mbox{CH}_{2}\mbox{CH}_{3}$  or  $\mbox{CH}_{2}\mbox{CH}_{2}\mbox{CH}_{2}\mbox{OH},$ 

R<sup>8</sup> is H or CH<sub>3</sub>, and

the sum of X and Y is 40 to 4000;

or a dispersed polyorganosiloxane which comprises at least one unit of the formula (5);

(5) 
$$(R^9)_V (R^{10})_W \text{Si-A-B}$$

wherein

R<sup>9</sup> is CH<sub>3</sub> or CH<sub>3</sub>CH<sub>2</sub>,

R<sup>10</sup> is -O-Si or -O-R<sup>9</sup>,

the sum of v and w equals 3, and v does not equal 3,

 $A = -CH_2CH(R^{11})(CH_2)_K$ 

B =

(6) 
$$-(R^{13})_{n}U^{1}$$
  $U^{2}-R^{14}$   $CH_{2}-C$   $R^{15}$   $R^{15}$   $R^{15}$ 

n is 1,  $U^{1} \text{ is CH,}$  k is 0 to 6,  $R^{11} \text{ is H or CH}_{3},$   $R^{13} \text{ is OOCN(butyl),}$   $R^{14} \text{ is H, linear C}_{1}\text{-C}_{4}\text{alkyl, phenyl,}$   $R^{15} \text{ is H or linear C}_{1}\text{-C}_{4}\text{alkyl, and}$   $U^{2} \text{ is N:}$ 

or a dispersed polyorganosiloxane of the formula (8);

(8) 
$$R^{17} - Si - O = CH_3 - CH_3 -$$

### wherein

 $R^3$  is as previously defined,  $R^{17}$  is OH,  $OR^{18}$  or  $CH_3$ ,  $R^{18}$  is  $CH_3$  or  $CH_2CH_3$ ,  $R^{19}$  is  $R^{20}$ - $(EO)_m$ - $(PO)_n$ - $R^{21}$ ,

m is 3 to 25,

n is 0 to 10,

 $R^{20}$  is the direct bond or  $CH_2CH(R^{22})(CH_2)_pR^{23}$ ,

p is 1 to 4,

 $R^{21}$  is H,  $R^{24}$ ,  $CH_2CH(R^{22})NH_2$  or  $CH(R^{22})CH_2NH_2$ ,

R<sup>22</sup> is H or CH<sub>3</sub>,

R<sup>23</sup> is O or NH,

 $R^{24}$  is linear or branched  $C_1$ - $C_3$ alkyl or  $Si(R^{25})_3$ ,

R<sup>25</sup> is R<sup>24</sup>, OCH<sub>3</sub> or OCH<sub>2</sub>CH<sub>3</sub>,

EO is -CH2CH2O- .

PO is -CH(CH<sub>3</sub>)CH<sub>2</sub>O- or -CH<sub>2</sub>CH(CH<sub>3</sub>)O- and

the sum of  $X_1, Y_1$  and S is 20 to 1500;

or a dispersed polyorganosiloxane of the formula (9);

(9) 
$$H_3C - Si - O = \begin{bmatrix} CH_3 & CH_3 &$$

wherein

R<sup>26</sup> is linear C<sub>1</sub>-C<sub>20</sub>alkoxy,

R<sup>4</sup> is as previously defined,

R<sup>29</sup> is linear C<sub>1</sub>-C<sub>20</sub>alkyl,

R<sup>27</sup> is CH<sub>2</sub>CH(R<sup>4</sup>)penyl,

R<sup>28</sup> is

(10) 
$$(CH_2)_3$$
— $O$ — $CH_2$ — $CH_2$ — $CH_2$ , and

the sum of  $X^2$ ,  $X^3$ ,  $X^4$  and  $Y^2$  is 20 to 1500, wherein  $X^3$ ,  $X^4$  and  $Y^2$  may be independently of each other 0;

or a mixture thereof.

23. (new) A method of use according to claim 21 wherein a polyorganosiloxane of formula (1) is used, wherein

R<sup>1</sup> is OH or CH<sub>3</sub>,

R<sup>3</sup> is CH<sub>3</sub>, C<sub>10</sub>-C<sub>20</sub>alkoxy or CH<sub>2</sub>CHR<sup>4</sup>CH<sub>2</sub>NHR<sup>5</sup>,

R⁴ is H,

R<sup>5</sup> is H or CH<sub>2</sub>CH<sub>2</sub>NHR<sup>6</sup>,

 $R^6$  is H or C(=0)- $R^7$ , and

R<sup>7</sup> is CH<sub>3</sub>, CH<sub>2</sub>CH<sub>3</sub> or CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>OH.

24. (new) A method of use according to claim 1 wherein a polyorganosiloxane of formula (8) is used, wherein

 $R^3$  is  $CH_3$ ,  $C_{10}$ - $C_{20}$ alkoxy or  $CH_2CHR^4CH_2NHR^5$ ,

R<sup>4</sup> is H,
R<sup>5</sup> is H or CH<sub>2</sub>CH<sub>2</sub>NHR<sup>6</sup>,
R<sup>6</sup> is H or C(=Q)-R<sup>7</sup>,
R<sup>7</sup> is CH<sub>2</sub>CH<sub>3</sub>, CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>OH or CH<sub>3</sub>, and
R<sub>17</sub> is CH<sub>3</sub> or OH.

25. (new) A method of use according to claim 21 wherein a polyorganosiloxane of formula (9) is used, wherein

R<sup>26</sup> is CH<sub>2</sub>CH(R<sup>4</sup>)R<sup>29</sup>,

R<sup>4</sup> is H, and

R<sup>27</sup> is 2-phenylpropyl.

- 26. (new) A method of use according to claim 21 wherein the composition is a liquid aqueous composition.
- 27. (new) A method of use according to claim 21 wherein the composition is used in a tumble dryer sheet composition.
- 28. (new) A method of use according to claim 21 in which the polyorganosiloxane is nonionic or cationic.
- 29. (new) A method of use according to claim 21 in which the composition has a solids content of 5 to 70 % at a temperature of 120°C.
- 30. (new) A method of use according to claim 21 in which the composition contains a water content of 25 to 90 % by weight based on the total weight of the composition.
- 31. (new) A method of use according to claim 21 in which the composition has a pH value from 2 to 7.
- 32. (new) A method of use according to claim 21 in which the nitrogen content of the aqueous emulsion due to the polyorganosiloxane is from 0 to 0.25 % with respect to the silicon content.
- 33. (new) A method of use according to claim 21 wherein the composition comprises a polyethylene, a fatty acid alkanolamide or a polyurethane.

- 34. (new) A method of use according to claim 21 wherein the composition comprises a polyethylene or a fatty acid alkanolamide.
- 35. (new) A method of use according to claim 21 wherein the composition comprises a fatty acid alkanolamide.
- 36. (new) A method of use according to claim 21 wherein the composition comprises a polyethylene.
- 37. (new) A method of use according to claim 21 wherein the composition is prepared by mixing a preformulated fabric softener with an emulsion comprising the polyorganosiloxane and the additive.
- 38. (new) A method of use according to claim 21 wherein composition has a clear appearance.
- 39. (new) A method of use according to claim 21 in which the composition comprises:
- a) 0.01 to 70 % by weight, based on the total weight of the composition, of a polyorganosiloxane, or a mixture thereof;
- b) 0.2 to 15 % by weight based on the total weight of an emulsifier, or a mixture thereof;
- c) 0.01 to 15 % by weight based on the total weight of at least one additive selected from the group consisting of a polyethylene, a fatty acid alkanolamide, a polysilicic acid and a polyurethane, and d) water to 100 %.
- 40. (new) A tumble dryer sheet comprising a composition as defined in claim 21.--

## **REMARKS**

Claims 21-40 are pending. Claims 1-20 have been replaced by added claims 21-40. Claims 3-19 were replaced to correct informalities and reduce filing fees by eliminating multiple dependency. Claim 1 was replaced to provide minor clarification. Claim 20 was replaced to change dependency.

Newly added claims 21-40 are supported by originally filed claims 1-20 and the disclosure at page 7, third full paragraph. No new matter has been added.

Applicants aver that the claims are now in proper form for examination. An Action on the merits of the claims is respectfully awaited.

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APR 03 2002

Respectfully submitted,

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10 Rec'd PCI/FTO 0 3 APR 2002 10/089852

FABRIC SOFTENER COMPOSITIONS

#### FIELD OF THE INVENTION

The present invention relates to the use of fabric softener compositions comprising selected polyorganosiloxanes, or mixtures thereof, together with selected additives for the improvement of hydrophilicity properties of textile materials in domestic applications. In particular it relates to textile softening compositions for use in a textile laundering operation to impart excellent hydrophilicity properties on the textile.

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### BACKGROUND OF THE INVENTION

The present invention relates to a method for increasing hydrophilicity of a fabric material. More particularly, the invention relates to a method for imparting a durably increased capacity of water absorption and a durably decreased susceptibility to accumulation of static electricity.

Needless to say, fabric materials currently on use both in the clothing use of people and in the industrial applications are in a very large part produced of synthetic fibers or traditional natural fibers. One of the largest differences between the properties of the synthetic and natural fibers is in the hydrophilicity-hydrophobicity behavior of them, the former fibers being of course outstandingly less hydrophilic than the latter. The remarkably small hydrophilicity of synthetic fibers sometimes causes serious problems not encountered in the use of natural fibers.

For example, fabric materials made of synthetic fibers have a very poor capacity of water or sweat absorption, which is advantageous on one hand but disadvantageous on the other, so that wearers of clothes made of synthetic fibers unavoidably have an unpleasant feeling of heavy stuffiness especially when the clothes are underwears worn in contact with or in the proximity of the skin of the wearer in a hot and humid climate.

Another serious problem caused by the poor hydrophilicity of synthetic fibers is the great accumulation of static electricity on the fibers causing unpleasantness to the wearer of clothes of synthetic fibers in such a charged condition.

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Many attempts have of course been proposed and practiced in the prior art to solve these problems by increasing the hydrophilicity of the fabric materials of synthetic fibers and also natural fibres. For example, the problem of poor water absorption of synthetic fibers can be mitigated by the mixed spinning or mixed weaving with water-absorptive natural fibers. The effectiveness of this method is, however, limited since too much amounts of the natural fibers mixed with the synthetic fibers to attain sufficient hydrophilicity of the fabric material naturally result in the loss of the advantages inherent to synthetic fibers. An alternative method is the treatment of the fabric material of synthetic fibers with a water-absorbent agent to impart hydrophilicity to the surface of the fibers. Extensive investigations have been and are being undertaken in this direction to propose various kinds of water-absorbent agents effective for a particular type of synthetic fibers. For example, the capacity of water absorption of polyester fibers, e.g. polyethylene terephthalate fibers, can be increased by the treatment with a water-soluble polyester resin. Unfortunately, such a method of the treatment of synthetic fibers with a water-soluble resin is defective in several respects of the poor durability of the effects obtained therewith and the adverse influences on the color fastness of dyed fabric materials in many cases.

Limiting the matter to the antistatic treatment or decrease of accumulation of static electricity on the synthetic fibers, various antistatic agents have been proposed hitherto. For example, the above mentioned water-soluble resins including water-soluble polyester resins, polyurethane resins, polyacrylamide resins, polyamide resins and the like are of course effective as an antistatic agent with certain durability. Besides, many compounds are known to be effective as an antistatic agent including inorganic salts such as calcium chloride and lithium chloride, guanidine compounds such as guanidine hydrochloride, surface active agents such as those of the types of quaternary ammonium salts and phosphoric acid esters, acrylic polymers having quaternary cationic groups and the like although the effectiveness of the treatment with these compounds is rather temporary.

The durability of the effects obtained with the above described antistatic agents is, however, not quite satisfactory even with the relatively durable polymeric antistatic agents and the antistatic effects obtained therewith are decreased in the long-run use of the treated fabric materials even by setting aside the other problem of the insufficient effectiveness of the method. Furthermore, the method is also not free from the problem of the decreased color

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fastness of dyed fabric materials giving limitations to the amount and the manner of use of the antistatic agents.

In short, none of the prior art methods by use of a hydrophilic agent, i.e. water-absorbent agent or antistatic agent, is quite satisfactory for imparting hydrophilicity to the fabric materials in respects of the effectiveness and the durability.

As given above one component of the compositions of the present invention are polyorganosiloxanes. Such compounds are known to be used on an industrial scale to finish fabrics by providing them with a permanent or semi-permanent finish aimed at improving their general appearance. Significant for these industrial fabric finishing processes is a co-called curing step generally involving temperatures in excess of 150°C often for periods of one hour or more. The object here is to form a chemical finish which resists destruction during subsequent cleaning/laundering of fabrics. This process of finishing is not carried out in domestic applications and accordingly one would not expect benefits of a comparable nature or magnitude from polyorganosiloxanes included as adjuncts in domestic softeners. Indeed, it is noteworthy that if the compounds of the current invention achieved a permanence associated with industrial textile finishing, problems associated with a cumulative build through the wash cycles could occur such as fabric discoloration and even in extremes an unpleasant feel to the wearer.

Surprisingly, it has been found that the use of selected polyorganosiloxanes, or mixtures thereof, and selected additives in fabric softener compositions provide excellent hydrophilic effects when applied to fabrics during a textile laundry operation.

Similar benefits are noted when compositions of the current invention are incorporated into tumble dryer additives such as impregnates on sheets.

## SUMMARY OF THE INVENTION

This invention relates to a method of use of a fabric softener composition for imparting hydrophilicity to textile fibre materials in domestic applications, which softener composition comprises:

A) a fabric softener;

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- B) at least one additive selected from the group consisting of
  - a) a polyethylene, or a mixture thereof,
  - b) a fatty acid alkanolamide, or a mixture thereof,
  - c) a polysilicic acid, or a mixture thereof, and
  - d) a polyurethane, or a mixture thereof; and
- C) a dispersed polyorganosiloxane of formula (1)

(1) 
$$R^{1} - S_{1} - O = \begin{bmatrix} CH_{3} \\ | \\ S_{1} - O \end{bmatrix} = \begin{bmatrix} CH_{3} \\ | \\ S_{1} - O \end{bmatrix} = \begin{bmatrix} CH_{3} \\ | \\ S_{1} - O \end{bmatrix} = \begin{bmatrix} CH_{3} \\ | \\ S_{1} - O \end{bmatrix} = \begin{bmatrix} CH_{3} \\ | \\ CH_{3} \end{bmatrix} = \begin{bmatrix} CH_{$$

wherein

R1 is OH, OR2 or CH3

R<sup>2</sup> is CH<sub>3</sub> or CH<sub>2</sub>CH<sub>3</sub>

R3 is C1-C20alkoxy, CH3, CH2CHR4CH2NHR5, or CH2CHR4CH2N(COCH3)R5

(2) 
$$(CH_2)_3O$$
 NR<sup>8</sup>

or (3)  $(CH_2)_3NH$  CH

or (4)  $(CH_2)_3$  NR<sup>8</sup>

R<sup>4</sup> is H or CH<sub>3</sub>

 $R^5$  is H,  $CH_2CH_2NHR^6$ ,  $C(=O)-R^7$  or  $(CH_2)_Z-CH_3$ 

z is 0 to 7

 $R^6$  is H or C(=O)- $R^7$ 

R<sup>7</sup> is CH<sub>3</sub>, CH<sub>2</sub>CH<sub>3</sub> or CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>OH

R<sup>8</sup> is H or CH<sub>3</sub>

the sum of X and Y is 40 to 4000;

or a dispersed polyorganosiloxane which comprises at least one unit of the formula (5)

(5) 
$$(R^9)_V (R^{10})_W \text{ Si-A-B}$$

wherein

R9 is CH3, CH3CH2 or Phenyl

the sum of v and w equals 3, and v does not equal 3

 $A = -CH_2CH(R^{11})(CH_2)_K$ 

 $B = -NR^{12}((CH_2)_I-NH)_mR^{12}$ , or

$$R^{15}$$
  $R^{15}$ 
 $CH_2$   $C$ 
 $-(R^{13})_{\overline{h}}U^{1}$   $U^2-R^{14}$ 
 $CH_2$   $C$ 
 $R^{15}$   $R^{15}$ 

n is 0 or 1

when n is 0, U1 is N, when n is 1, U1 is CH

I is 2 to 8

k is 0 to 6

m is 0 to 3

R<sup>11</sup> is H or CH<sub>3</sub>

 $R^{12}$  is H, C(=O)- $R^{16}$ ,  $CH_2(CH_2)_pCH_3$  or

p is 0 to 6

R<sup>13</sup> is NH, O, OCH<sub>2</sub>CH(OH)CH<sub>2</sub>N(Butyl), OOCN(Butyl)

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R<sup>14</sup> is H, linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl, Phenyl or CH<sub>2</sub>CH(OH)CH<sub>3</sub> R<sup>15</sup> is H or linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl R<sup>16</sup> is CH<sub>3</sub>, CH<sub>2</sub>CH<sub>3</sub> or (CH<sub>2</sub>)<sub>q</sub>OH

q is 1 to 6

U<sup>2</sup> is N or CH;

or a dispersed polyorganosiloxane of the formula (8)

(8) 
$$R^{17} - Si - O = CH_3 - CH_3 -$$

### wherein

R<sup>3</sup> is as previously defined

R<sup>17</sup> is OH, OR<sup>18</sup> or CH<sub>3</sub>

R<sup>18</sup> is CH<sub>3</sub> or CH<sub>2</sub>CH<sub>3</sub>

 $R^{19}$  is  $R^{20}$ -(EO)<sub>m</sub>-(PO)<sub>n</sub>- $R^{21}$ 

m is 3 to 25

n is 0 to 10

R<sup>20</sup> is the direct bond or CH<sub>2</sub>CH(R<sup>22</sup>)(CH<sub>2</sub>)<sub>p</sub>R<sup>23</sup>

p is 1 to 4

R<sup>21</sup> is H, R<sup>24</sup>, CH<sub>2</sub>CH(R<sup>22</sup>)NH<sub>2</sub> or CH(R<sup>22</sup>)CH<sub>2</sub>NH<sub>2</sub>

R<sup>22</sup> is H or CH<sub>3</sub>

R<sup>23</sup> is O or NH

R<sup>24</sup> is linear or branched C<sub>1</sub>-C<sub>8</sub> alkyl or Si(R<sup>25</sup>)<sub>3</sub>

R<sup>25</sup> is R<sup>24</sup>, OCH<sub>3</sub> or OCH<sub>2</sub>CH<sub>3</sub>

EO is -CH2CH2O-

PO is -CH(CH<sub>3</sub>)CH<sub>2</sub>O- or -CH<sub>2</sub>CH(CH<sub>3</sub>)O-

the sum of  $X_1, Y_1$  and S is 20 to 1500;

or a dispersed polyorganosiloxane of the formula (9)

### wherein

R<sup>26</sup> is linear or branched C<sub>1</sub> - C<sub>20</sub> alkoxy, CH<sub>2</sub>CH(R<sup>4</sup>)R<sup>29</sup>

R<sup>4</sup> is as previously defined

R<sup>29</sup> is linear or branched C<sub>1</sub> - C<sub>20</sub> alkyl

 $R^{27}$  is aryl, aryl substituted by linear or branched  $C_1$  -  $C_{10}$  alkyl, linear or branched  $C_1$  -  $C_{20}$  alkyl substituted by aryl or aryl substituted by linear or branched  $C_1$  -  $C_{10}$  alkyl  $R^{28}$  is

the sum of  $X^2$ ,  $X^3$ ,  $X^4$  and  $Y^2$  is 20 to 1500, wherein  $X^3$ ,  $X^4$  and  $Y^2$  may be independently of each other 0;

or a mixture thereof.

The composition is preferably used as a liquid rinse conditioner composition. The textile fibre materials are treated for hydrophilicity.

In tumble dryer applications the compositions are usually incorporated into impregnates on non-woven sheets. However, other application forms are known to those skilled in the art.

The fabric softener composition (especially in liquid form) will be used after the textile fibre materials have been washed with a laundry detergent, which may be one of a broad range of detergent types. The tumble dryer sheet will be used after a laundering process. The textile fibre materials may be damp or dry.

The fabric softener composition may also be sprayed directly onto the fabrics prior to or during the ironing or drying of the treated fabrics.

The polyorganosiloxane may be anionic, nonionic or cationic, preferably nonionic or cationic.

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The polyorganosiloxanes, or mixtures thereof, are used in a dispersed form, via the use of an emulsifier. The fabric softener composition is preferably in aqueous liquid form. The water content as a rule is 25 to 90% by weight based on the total weight of the composition.

When the polyorganosiloxane contains a nitrogen atom, the nitrogen content of the aqueous emulsion due to the polyorganosiloxane is preferably from 0.001 to 0.25 % with respect to the silicon content. In general, a nitrogen content from 0.001 to 0.25 % is preferred. The particles of the emulsion as a rule have a diameter of between 5nm and 1000nm.

The fabric softener composition preferably has a solids content of 5 to 70% at a temperature of 120°C.

The fabric softener composition usually has a pH value from 2.0 to 7.0, especially 2.0 to 5.0.

The fabric softener composition may further comprise an additional polyorganosiloxane:

wherein g is

and G is C<sub>1</sub> to C<sub>20</sub> alkyl.

This polydimethylsiloxane is cationic, has a viscosity at 25°C of 250 mm<sup>2</sup>s<sup>-1</sup> to 450 mm<sup>2</sup>s<sup>-1</sup>, has a specific gravity of 1.00 to 1.02 g/cm<sup>3</sup> and has a surface tension of 28.5 mNm<sup>-1</sup> to 33.5 mNm<sup>-1</sup>.

The fabric softener composition may further comprise an additional polyorganosiloxane, such as that known as Magnasoft HSSD, or a polyorganosiloxane of the formula:

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(13) 
$$H_3C - Si - O = CH_3$$

$$CH_3$$

$$CH_3$$

$$Si - O = Si - CH_3$$

$$CH_3$$

$$Si - O = Si - CH_3$$

$$CH_3$$

R is CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>N(R)<sub>2</sub>
R is linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl
R is (CH<sub>2</sub>)<sub>X''</sub>-(EO)<sub>m</sub>-(PO)<sub>n</sub>-R
m is 3 to 25
n is 0 to 10
X is 0 to 4
R''' is H or linear or branched C<sub>1</sub>-C<sub>4</sub> alkyl
EO is -CH<sub>2</sub>CH<sub>2</sub>OPO is -CH(CH<sub>3</sub>)CH<sub>2</sub>O- or -CH<sub>2</sub>CH(CH<sub>3</sub>)Othe sum of X , Y and S is 40 to 300.

Preferably the compositions comprise dispersed polyorganosiloxanes of formula (1):

(1) 
$$R^{1} - S_{1} - O = \begin{bmatrix} CH_{3} \\ | \\ S_{1} - O \end{bmatrix} = \begin{bmatrix} CH_{3} \\ | \\ S_{1} - O \end{bmatrix} = \begin{bmatrix} CH_{3} \\ | \\ S_{1} - O \end{bmatrix} = \begin{bmatrix} CH_{3} \\ | \\ S_{1} - O \end{bmatrix} = \begin{bmatrix} CH_{3} \\ | \\ S_{1} - O \end{bmatrix} = \begin{bmatrix} CH_{3} \\ | \\ CH_{3} \end{bmatrix} = \begin{bmatrix} C$$

## wherein

R<sup>1</sup> is OH, OR<sup>2</sup> or CH<sub>3</sub>
R<sup>2</sup> is CH<sub>3</sub> or CH<sub>2</sub>CH<sub>3</sub>
R<sup>3</sup> is C₁-C<sub>20</sub>alkoxy, CH<sub>3</sub>, CH<sub>2</sub>CHR<sup>4</sup>CH<sub>2</sub>NHR<sup>5</sup>, or

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(2) 
$$(CH_2)_3O$$
 NR<sup>8</sup>
or (3)  $(CH_2)_3NH$  CH

R⁴ is H or CH₃

 $R^5$  is H,  $CH_2CH_2NHR^6$ ,  $C(=0)-R^7$ 

 $R^6$  is H or C(=O)- $R^7$ 

R<sup>7</sup> is CH<sub>3</sub>, CH<sub>2</sub>CH<sub>3</sub> or CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>OH

R<sup>8</sup> is H or CH<sub>3</sub>

the sum of X and Y is 40 to 4000, especially 40 to 2000;

or a dispersed polyorganosiloxane which comprises at least one unit of the formula (5);

(5) 
$$(R^9)_V (R^{10})_W \text{Si-A-B}$$

wherein

R<sup>9</sup> is CH<sub>3</sub>, CH<sub>3</sub>CH<sub>2</sub>

R<sup>10</sup> is -O-Si or -O-R<sup>9</sup>

the sum of v and w equals 3, and v does not equal 3

 $\mathsf{A} = -\mathsf{CH}_2\mathsf{CH}(\mathsf{R}^{11})(\mathsf{CH}_2)_\mathsf{K}$ 

B =

(6)

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n is 1

U1 is CH

k is 0 to 6

R<sup>11</sup> is H or CH<sub>3</sub>

R<sup>13</sup> is OOCN(Butyl)

R<sup>14</sup> is H, linear C<sub>1</sub>-C<sub>4</sub> alkyl, Phenyl

R<sup>15</sup> is H or linear C<sub>1</sub>-C<sub>4</sub> alkyl

U<sup>2</sup> is N;

or a dispersed polyorganosiloxane of the formula (8);

(8) 
$$R^{17} - S_{1} - O = CH_{3} - CH_$$

## wherein

R<sup>3</sup> is as previously defined

R<sup>17</sup> is OH, OR<sup>18</sup> or CH<sub>3</sub>

R<sup>18</sup> is CH<sub>3</sub> or CH<sub>2</sub>CH<sub>3</sub>

 $R^{19}$  is  $R^{20}$ -(EO)<sub>m</sub>-(PO)<sub>n</sub>- $R^{21}$ 

m is 3 to 25

n is 0 to 10

R<sup>20</sup> is the direct bond or CH<sub>2</sub>CH(R<sup>22</sup>)(CH<sub>2</sub>)<sub>0</sub>R<sup>23</sup>

p is 1 to 4

R<sup>21</sup> is H, R<sup>24</sup>, CH<sub>2</sub>CH(R<sup>22</sup>)NH<sub>2</sub> or CH(R<sup>22</sup>)CH<sub>2</sub>NH<sub>2</sub>

R<sup>22</sup> is H or CH<sub>3</sub>

R<sup>23</sup> is O or NH

R<sup>24</sup> is linear or branched C<sub>1</sub>-C<sub>3</sub> alkyl or Si(R<sup>25</sup>)<sub>3</sub>

R<sup>25</sup> is R<sup>24</sup>, OCH<sub>3</sub> or OCH<sub>2</sub>CH<sub>3</sub>

EO is -CH2CH2O-

PO is -CH(CH<sub>3</sub>)CH<sub>2</sub>O- or -CH<sub>2</sub>CH(CH<sub>3</sub>)O-

the sum of X<sup>1</sup>,Y<sup>1</sup> and s is 20 to 1500;

or a dispersed polyorganosiloxane of the formula (9);

$$(9) \quad H_{3}C = \begin{bmatrix} CH_{3} & CH$$

R<sup>26</sup> is linear C<sub>1</sub> - C<sub>20</sub> alkoxy,

R4 is as previously defined

R<sup>29</sup> is linear C<sub>1</sub> - C<sub>20</sub> alkyl

R<sup>27</sup> is, CH<sub>2</sub>CH(R<sup>4</sup>)Phenyl

R<sup>28</sup> is

(10) 
$$(CH_2)_3$$
  $-CH_2$   $-CH_2$   $-CH_2$   $O$ 

the sum of  $X^2$ ,  $X^3$ ,  $X^4$  and  $Y^2$  is 20 to 1500, wherein  $X^3$ ,  $X^4$  and  $Y^2$  may be independently of each other 0;

or a mixture thereof.

The fabric softener composition may further comprise an additional polyorganosiloxane:

wherein g is

$$(12) \begin{array}{c} OH \\ | \\ ---CH_2--CH----CH_2--O-(CH_2)_3---- \end{array}$$

and G is C<sub>1</sub> to C<sub>20</sub> alkyl.

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This polydimethylsiloxane is cationic, has a viscosity at 25°C of 250 mm<sup>2</sup>s<sup>-1</sup> to 450 mm<sup>2</sup>s<sup>-1</sup>, has a specific gravity of 1.00 to 1.02 g/cm<sup>3</sup> and has a surface tension of 28.5 mNm<sup>-1</sup> to 33.5 mNm<sup>-1</sup>.

As to the polyorganosiloxanes of formula (1) the following preferences apply:

R<sup>1</sup> is preferably OH or CH<sub>3</sub>.

R<sup>3</sup> is preferably CH<sub>3</sub>, C<sub>10</sub>-C<sub>20</sub>alkoxy or CH<sub>2</sub>CHR<sup>4</sup>CH<sub>2</sub>NHR<sup>5</sup>.

R<sup>4</sup> is preferably H.

R<sup>5</sup> is preferably H or CH<sub>2</sub>CH<sub>2</sub>NHR<sup>6</sup>.

 $R^6$  is preferably H or C(=0)- $R^7$ .

R<sup>7</sup> is preferably CH<sub>3</sub>, CH<sub>2</sub>CH<sub>3</sub> or especially CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>OH.

The sum of X + Y is preferably 100 to 2000.

Preferred are polyorganosiloxanes of formula (1) wherein

R1 is OH or CH3,

R<sup>3</sup> is CH<sub>3</sub>, C<sub>10</sub>-C<sub>20</sub>alkoxy or CH<sub>2</sub>CHR<sup>4</sup>CH<sub>2</sub>NHR<sup>5</sup>,

R⁴ is H.

R<sup>5</sup> is H or CH<sub>2</sub>CH<sub>2</sub>NHR<sup>6</sup>,

 $R^6$  is H or C(=0)- $R^7$ , and

R<sup>7</sup> is CH<sub>3</sub>, CH<sub>2</sub>CH<sub>3</sub> or especially CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>OH.

As to the polyorganosiloxanes of formula (8) the following preferences apply:

R<sup>3</sup> is preferably CH<sub>3</sub>, C<sub>10</sub>-C<sub>20</sub>alkoxy or CH<sub>2</sub>CHR<sup>4</sup>CH<sub>2</sub>NHR<sup>5</sup>.

R⁴ is preferably H.

R<sup>5</sup> is preferably H or CH<sub>2</sub>CH<sub>2</sub>NHR<sup>6</sup>.

R<sup>6</sup> is preferably H or C(=0)-R<sup>7</sup>.

R<sup>7</sup> is preferably CH<sub>2</sub>CH<sub>3</sub>, CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>OH or especially CH<sub>3</sub>.

R<sub>17</sub> is preferably CH<sub>3</sub> or OH.

R<sub>20</sub> is preferably the direct bond.

R<sub>21</sub> is preferably H.

Preferred are polyorganosiloxanes of formula (8) wherein

R<sup>3</sup> is CH<sub>3</sub>, C<sub>10</sub>-C<sub>20</sub>alkoxy or CH<sub>2</sub>CHR<sup>4</sup>CH<sub>2</sub>NHR<sup>5</sup>,

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 $R^4$  is H,  $R^5$  is H or  $CH_2CH_2NHR^6$ ,  $R^6$  is H or  $C(=O)-R^7$ ,  $R^7$  is  $CH_2CH_3$ ,  $CH_2CH_2CH_2OH$  or especially  $CH_3$ , and  $R_{17}$  is  $CH_3$  or OH.

As to the polyorganosiloxanes of formula (9) the following preferences apply:  $R^{26}$  is preferably  $CH_2CH(R^4)R^{29}$ .  $R^4$  is preferably H.  $R^{27}$  is preferably 2-phenyl propyl. The sum of  $X^2$ ,  $X^3$ ,  $X^4$  and  $Y^2$  is preferably 40 to 500.

Preferred are polyorganosiloxanes of formula (9) wherein R<sup>26</sup> is CH<sub>2</sub>CH(R<sup>4</sup>)R<sup>29</sup>, R<sup>4</sup> is H, and R<sup>27</sup> is 2-phenyl propyl.

Preferred are polyorganosiloxanes of formulae (1), (8) and (9), especially those of formulae (1) and (8). Very interesting polyorganosiloxanes are those of formula (1).

Emulsifiers used to prepare the polyorganosiloxane compositions include:

- i) Ethoxylates, such as alkyl ethoxylates, amine ethoxylates or ethoxylated alkylammoniumhalides. Alkyl ethoxylates include alcohol ethoxylates or isotridecyl ethoxylates. Preferred alcohol ethoxylates include linear or branched nonionic alkyl ethoxylates containing 2 to 15 ethylene oxide units. Preferred isotridecyl ethoxylates include nonionic isotridecyl ethoxylates containing 5 to 25 ethylene oxide units. Preferred amine ethoxylates include nonionic C10 to C20 alkyl amino ethoxylates containing 4 to 10 ethylene oxide units. Preferred ethoxylated alkylammoniumhalides include nonionic or cationic ethoxylated C6 to C20 alkyl bis(hydroxyethyl)methylammonium chlorides.
- ii) Alkylammonium halides, preferably cationic quaternary ester alkylammonium halides.
- iii) Silicones, preferably nonionic polydimethylsiloxane polyoxyalkylene copolymers
- iv) Saccharides, preferably nonionic alkylpolyglycosides.

A mixture of these emulsifiers may also be used.

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As mentioned previously, the compositions further comprise one or more additives selected from polyethylene, dispersed fatty acid alkanol amide, polysilicic acid and polyurethane. These components are described below.

The emulsifiable polyethylene (polyethylene wax) is known and is described in detail in the prior art (compare, for example, DE-C-2,359,966, DE-A-2,824,716 and DE-A-1,925,993). The emulsifiable polyethylene is as a rule a polyethylene having functional groups, in particular COOH groups, some of which can be esterified. These functional groups are introduced by oxidation of the polyethylene. However, it is also possible to obtain the functionality by copolymerization of ethylene with, for example, acrylic acid. The emulsifiable polyethylenes preferably have a density of at least 0.91 g/cm³ at 20°C., an acid number of at least 5 and a saponification number of at least 10. Emulsifiable polyethylenes which have a density of 0.95 to 1.05 g/cm³ at 20°C, an acid number of 10 to 60 and a saponification number of 15 to 80 are particularly preferred. Polyethylenes which have a drop point of 100-150°C are preferred. This material is generally obtainable commercially in the form of flakes, lozenges and the like. A mixture of these emulsifiable polyethylenes may also be used.

The polyethylene wax is usually employed in the form of dispersions. Various emulsifiers are suitable for their preparation. The preparation of the dispersions is described in detail in the prior art.

Emulsifiers suitable for dispersing the polyethylene component include:

- i) Ethoxylates, such as alkyl ethoxylates or amine ethoxylates. Alkyl ethoxylates include alcohol ethoxylates or isotridecyl ethoxylates. Preferred alcohol ethoxylates include nonionic fatty alcohol ethoxylates containing 2 to 55 ethylene oxide units. Preferred isotridecyl ethoxylates include nonionic isotridecyl ethoxylates containing 6 to 9 ethylene oxide units. Preferred amine ethoxylates include nonionic C10 to C20 alkyl amino ethoxylates containing 7 to 9 ethylene oxide units.
- ii) Alkylammonium halides, preferably cationic quaternary ester alkylammonium halides.
- iii) Ammonium salts, preferably cationic aliphatic quaternary ammonium chloride or sulfate.

A mixture of these emulsifiers may also be used.

Suitable fatty acid alkanolamides are for example those of formula

(14) 
$$R_{33} C N R_{34}$$

#### wherein

R<sub>33</sub> is a saturated or unsaturated hydrocarbon radical containing 10 to 24 carbon atoms,

number from 1 to 10 and R<sub>36</sub> is as defined above for R<sub>33</sub>, and

 $R_{35}$  is a radical of formula -CH<sub>2</sub>OH, -(CH<sub>2</sub>CH<sub>2</sub>O)<sub>c</sub>H, —CH<sub>2</sub>CH<sub>2</sub>—N or  $R_{37}$ 

c is as defined above,

 $R_{38}$ ,  $R_{38}$ ' and  $R_{38}$ " have the same or different meaning and are as defined above for  $R_{34}$ , and  $R_{39}$ ,  $R_{39}$ ' and  $R_{39}$ " have the same or different meaning and are a radical of formula

R<sub>33</sub> and R<sub>36</sub> are preferably a saturated or unsaturated hydrocarbon radical containing 14 to 24 carbon atoms. Preferred are saturated hydrocarbon radicals.

R<sub>35</sub> is preferably a radical of formula

As to  $R_{38}$ ,  $R_{38}$ ' and  $R_{38}$ " the preferences given above for  $R_{34}$  apply.

c is preferably a number from 1 to 5.

Preferred are fatty acid alkanolamides of formula

$$\begin{array}{c} & \text{P}_{33} & \text{P}_{34} \\ & \text{P}_{33} & \text{P}_{34} \\ & \text{P}_{33} & \text{P}_{34} \\ & \text{P}_{34} & \text{P}_{34} \\ & \text{P}_{34} & \text{P}_{34} \\ & \text{P}_{35} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{39} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{39} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{39} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} & \text{P}_{36} \\ & \text$$

wherein R<sub>33</sub>, R<sub>34</sub>, R<sub>38</sub>, R<sub>38</sub>', R<sub>38</sub>", R<sub>39</sub>, R<sub>39</sub>' and R<sub>39</sub>" are as defined above.

Preferred are fatty acid alkanolamides of formula (15a), wherein R<sub>34</sub>, R<sub>38</sub>, R<sub>38</sub> and R<sub>38</sub> are hydrogen or -CH<sub>2</sub>OH.

Furthermore, fatty acid alkanolamides of formula

(15b) 
$$R_{33} = C - N R_{34} - (CH_2CH_2O)_CH$$

are preferred, wherein R<sub>33</sub>, R<sub>34</sub>, R<sub>37</sub> and c are as defined above.

Preferred are fatty acid alkanolamides of formula (15b), wherein

 $R_{34}$  and  $R_{37}$  are hydrogen or a radical of formula  $\begin{array}{c} O \\ \vdots \\ C-R_{2c} \end{array}$ .  $R_{34}$  is preferably hydrogen.

The above fatty acid alkanolamides can also be present in form of the corresponding ammonium salts.

A mixture of these fatty acid alkanolamides may also be used.

Emulsifiers suitable for dispersing the fatty acid alkanol amide component include:

- i) Ethoxylates, such as alkyl ethoxylates, amine ethoxylates or amide ethoxylates. Alkyl ethoxylates include alcohol ethoxylates or isotridecyl ethoxylates. Preferred alcohol ethoxylates include nonionic fatty alcohol ethoxylates containing 2 to 55 ethylene oxide units. Preferred isotridecyl ethoxylates include nonionic isotridecyl ethoxylates containing 5 to 45 ethylene oxide units. Preferred amine ethoxylates include nonionic C10 to C20 alkyl amino ethoxylates containing 4 to 25 ethylene oxide units. Preferred amide ethoxylates include cationic fatty acid amide ethoxylates containing 2 to 25 ethylene oxide units.
- ii) Alkylammonium halides, preferably cationic quaternary ester alkylammonium halides or cationic aliphatic acid alkylamidotrialkylammonium methosulfates.
- iii) Ammonium salts, preferably cationic aliphatic quaternary ammonium chloride or sulfate.

A mixture of these emulsifiers may also be used.

Examples for polyurethanes are the reaction products of a diol and an ethoxysilate with a diisocyanate.

The additives selected from the group consisting of a polyethylene, a fatty acid alkanolamide, a polysilicic acid, and a polyurethane are, as a rule, used in an amount of 0.01 to 25 % by weight, especially 0.01 to 15 % by weight, based on the total weight of the fabric

softener composition. An amount of 0.05 to 15 % by weight, especially 0.1 to 15 % by weight, is preferred. Highly preferred is an upper limit of 10 %, especially 5 %.

Preferred as additives are polyethylene, fatty acid alkanolamides and polyurethanes, especially polyethylene and fatty acid alkanolamides. Highly preferred are polyethylene.

A highly preferred fabric softener composition used according to the present invention comprises:

- a) 0.01 to 70 % by weight based on the total weight of the composition of a polyorganosiloxane, or a mixture thereof;
- b) 0.2 to 15 % by weight based on the total weight of an emulsifier, or a mixture thereof;
- c) 0.01 to 25 % by weight, especially 0.01 to 15 % by weight, based on the total weight of at least one additive selected from the group consisting of a polyethylene, a fatty acid alkanolamide, a polysilicic acid, or a polyurethane, and d) water to 100 %.

The fabric softener compositions can be prepared as follows:

Firstly, emulsions of the polyorganosiloxane are prepared. The polyorganosiloxane and polyethylene, fatty acid alkanol amide, polysilicic acid or polyurethane are emulsified in water using one or more surfactants and shear forces, e.g. by means of a colloid mill. Suitable surfactants are described above. The components may be emulsified individually before being mixed together, or emulsified together after the components have been mixed. The surfactant(s) is/are used in customary amounts known to the person skilled in the art and can be added either to the polyorganosiloxane or to the water prior to emulsification. Where appropriate, the emulsification operation can be carried out at elevated temperature. The fabric softener composition according to the invention is usually, but not exclusively, prepared by firstly stirring the active substance, i.e. the hydrocarbon based fabric softening component, in the molten state into water, then, where required, adding further desired additives and, finally, after cooling, adding the polyorganosiloxane emulsion.

The fabric softener composition can, for example, be prepared by mixing a preformulated fabric softener with an emulsion comprising the polyorganosiloxane and the additive.

The fabric softening components can be conventional hydrocarbon based fabric softening components known in the art.

Hydrocarbon fabric softeners suitable for use herein are selected from the following classes of compounds:

(i) Cationic quaternary ammonium salts. The counter ion of such cationic quaternary ammonium salts may be a halide, such as chloride or bromide, methyl sulphate, or other ions well known in the literature. Preferably the counter ion is methyl sulfate or any alkyl sulfate or any halide, methyl sulfate being most preferred for the dryer-added articles of the invention.

Examples of cationic quaternary ammonium salts include but are not limited to:

(1) Acyclic quaternary ammonium salts having at least two C<sub>8</sub> to C<sub>30</sub>, preferably C<sub>12</sub> to C<sub>22</sub> alkyl or alkenyl chains, such as: ditallowdimethyl ammonium methylsulfate, di(hydrogenated tallow)dimethyl ammonium methylsulfate, distearyldimethyl ammonium methylsulfate, dicocodimethyl ammonium methylsulfate and the like. It is especially preferred if the fabric softening compound is a water insoluble quaternary ammonium material which comprises a compound having two C<sub>12</sub> to C<sub>18</sub> alkyl or alkenyl groups connected to the molecule via at least one ester link. It is more preferred if the quaternary ammonium material has two ester links present. An especially preferred ester-linked quaternary ammonium material for use in the invention can be represented by the formula:

(16) 
$$R^{31}$$
  $\downarrow^{-}$   $\downarrow^{-}$   $(CH_2)e - T - R^{32}$   $(CH_2)e - T - R^{32}$ 

wherein each  $R^{31}$  group is independently selected from  $C_1$  to  $C_4$  alkyl, hydroxyalkyl or  $C_2$  to  $C_4$  alkenyl groups; T is either

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and wherein each  $R^{32}$  group is independently selected from  $C_8$  to  $C_{28}$  alkyl or alkenyl groups; and e is an integer from 0 to 5.

A second preferred type of quaternary ammonium material can be represented by the formula:

A second preferred type of quaternary ammonium material can be represented by the formula:

(19) 
$$(R^{31})_3N^+$$
— $(CH_2)_e$  —  $CH_2OOR^{32}$ 

wherein  $R^{31}$ , e and  $R^{32}$  are as defined above.

- (2) Cyclic quaternary ammonium salts of the imidazolinium type such as di(hydrogenated tallow)dimethyl imidazolinium methylsulfate, 1-ethylene-bis(2-tallow-1-methyl) imidazolinium methylsulfate and the like;
- (3) Diamido quaternary ammonium salts such as: methyl-bis(hydrogenated tallow amidoethyl)-2-hydroxethyl ammonium methyl sulfate, methyl bi(tallowamidoethyl)-2-hydroxypropyl ammonium methylsulfate and the like;
- (4) Biodegradable quaternary ammonium salts such as N,N-di(tallowoyl-oxy-ethyl)-N,N-dimethyl ammonium methyl sulfate and N,N-di(tallowoyl-oxy-propyl)-N,N-dimethyl ammonium methyl sulfate. Biodegradable quaternary ammonium salts are described, for

example, in U.S. Patents 4,137,180, 4,767,547 and 4,789,491 incorporated by reference herein.

Preferred biodegradable quaternary ammonium salts include the biodegradable cationic diester compounds as described in U.S. Patent 4,137,180, herein incorporated by reference.

- (ii) Tertiary fatty amines having at least one and preferably two C8 to C30, preferably C12 to C22 alkyl chains. Examples include hardened tallow-di-methylamine and cyclic amines such as 1-(hydrogenated tallow)amidoethyl-2-(hydrogenated tallow) imidazoline. Cyclic amines which may be employed for the compositions herein are described in U.S. Patent 4,806,255 incorporated by reference herein.
- (iii) Carboxylic acids having 8 to 30 carbons atoms and one carboxylic group per molecule. The alkyl portion has 8 to 30, preferably 12 to 22 carbon atoms. The alkyl portion may be linear or branched, saturated or unsaturated, with linear saturated alkyl preferred. Stearic acid is a preferred fatty acid for use in the composition herein. Examples of these carboxylic acids are commercial grades of stearic acid and palmitic acid, and mixtures thereof which may contain small amounts of other acids.
- (iv) Esters of polyhydric alcohols such as sorbitan esters or glycerol stearate. Sorbitan esters are the condensation products of sorbitol or iso-sorbitol with fatty acids such as stearic acid. Preferred sorbitan esters are monoalkyl. A common example of sorbitan ester is SPAN 60 (ICI) which is a mixture of sorbitan and isosorbide stearates.
- (v) Fatty alcohols, ethoxylated fatty alcohols, alkyphenols, ethoxylated alkyphenols, ethoxylated fatty amines, ethoxylated monoglycerides and ethoxylated diglycerides.
- (vi) Mineral oils, and polyols such as polyethylene glycol.

These softeners are more definitively described in U.S. Patent 4,134,838 the disclosure of which is incorporated by reference herein. Preferred fabric softeners for use herein are acyclic quaternary ammonium salts. Di(hydrogenated)tallowdimethyl ammonium methylsulfate is most widely used for dryer articles of this invention. Mixtures of the above mentioned fabric softeners may also be used.

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The fabric softening composition employed in the present invention contains as a rule about 0.1% to about 95% of the fabric softening component. Preferably from about 2% to about 70% and most preferably from about 2% to about 30% of the fabric softening component is employed herein to obtain optimum softening at minimum cost. When the fabric softening component includes a quaternary ammonium salt, the salt is used in the amount of about 2% to about 70%, preferably about 2% to about 30%.

The fabric softener composition may also comprise additives which are customary for standard commercial liquid rinse conditioners, for example alcohols, such as ethanol, n-propanol, i-propanol, polyhydric alcohols, for example glycerol and propylene glycol; amphoteric and nonionic surfactants, for example carboxyl derivatives of imidazole, oxyethylated fatty alcohols, hydrogenated and ethoxylated castor oil, alkyl polyglycosides, for example decyl polyglucose and dodecylpolyglucose, fatty alcohols, fatty acid esters, fatty acids, ethoxylated fatty acid glycerides or fatty acid partial glycerides; also inorganic or organic salts, for example water-soluble potassium, sodium or magnesium salts, non-aqueous solvents, pH buffers, perfumes, dyes, hydrotropic agents, antifoams, anti redeposition agents, polymeric or other thickeners, enzymes, optical brighteners, antishrink agents, stain removers, germicides, fungicides, antioxidants and corrosion inhibitors.

These fabric softener compositions are traditionally prepared as dispersions containing for example up to 20 % by weight of active material in water. They have a turbid appearance. However, alternative formulations usually containing actives at levels of 5 to 40 % along with solvents can be prepared as microemulsions which have a clear appearance (as to the solvents and the formulations see for example US-A-5,543,067 und WO-A-98/17757). The additives and polyorganosiloxanes of the present invention can be used for such compositions although it will be necessary to use them in microemulsion form to preserve the clear appearance of the fabric softener compositions which are microemulsions.

Another aspect of the invention is a tumble dryer sheet article. The fabric softener composition of the present invention may be coated onto a flexible substrate which carries a fabric conditioning amount of the composition and is capable of releasing the composition at dryer operating temperatures. The conditioning composition in turn has a preferred melting (or softening) point of about 25°C to about 150°C.

The fabric softener composition which may be employed in the invention is coated onto a dispensing means which effectively releases the fabric conditioning composition in a tumble dryer. Such dispensing means can be designed for single usage or for multiple uses. One such multi-use article comprises a sponge material releasably enclosing enough of the conditioning composition to effectively impart fabric softness during several drying cycles. This multi-use article can be made by filling a porous sponge with the composition. In use, the composition melts and leaches out through the pores of the sponge to soften and condition fabrics. Such a filled sponge can be used to treat several loads of fabrics in conventional dryers, and has the advantage that it can remain in the dryer after use and is not likely to be misplaced or lost.

Another article comprises a cloth or paper bag releasably enclosing the composition and sealed with a hardened plug of the mixture. The action and heat of the dryer opens the bag and releases the composition to perform its softening.

A highly preferred article comprises the inventive compositions releasably affixed to a flexible substrate such as a sheet of paper or woven or non-woven cloth substrate. When such an article is placed in an automatic laundry dryer, the heat, moisture, distribution forces and tumbling action of the dryer removes the composition from the substrate and deposits it on the fabrics.

The sheet conformation has several advantages. For example, effective amounts of the compositions for use in conventional dryers can be easily absorbed onto and into the sheet substrate by a simple dipping or padding process. Thus, the end user need not measure the amount of the composition necessary to obtain fabric softness and other benefits. Additionally, the flat configuration of the sheet provides a large surface area which results in efficient release and distribution of the materials onto fabrics by the tumbling action of the dryer.

The substrates used in the articles can have a dense, or more preferably, open or porous structure. Examples of suitable materials which can be used as substrates herein include paper, woven cloth, and non-woven cloth. The term "cloth" herein means a woven or non-woven substrate for the articles of manufacture, as distinguished from the term "fabric"

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which encompasses the clothing fabrics being dried in an automatic dryer.

It is known that most substances are able to absorb a liquid substance to some degree; however, the term "absorbent", as used herein, is intended to mean a substrate with an absorbent capacity (i.e., a parameter representing a substrates ability to take up and retain a liquid) from 4 to 12, preferably 5 to 7 times its weight of water.

If the substrate is a foamed plastics material, the absorbent capacity is preferably in the range of 15 to 22, but some special foams can have an absorbent capacity in the range from 4 to 12.

Determination of absorbent capacity values is made by using the capacity testing procedures described in U.S. Federal Specifications (UU-T-595b), modified as follows:

- 1. tap water is used instead of distilled water;
- 2. the specimen is immersed for 30 seconds instead of 3 minutes;
- 3. draining time is 15 seconds instead of 1 minute; and
- 4. the specimen is immediately weighed on a torsion balance having a pan with turned-up edges.

Absorbent capacity values are then calculated in accordance with the formula given in said Specification. Based on this test, one-ply, dense bleached paper (e.g., Kraft or bond having a basis weight of about 32 pounds per 3,000 square feet) has an absorbent capacity of 3.5 to 4; commercially available household one-ply towel paper has a value of 5 to 6; and commercially available two-ply household towelling paper has a value of 7 to about 9.5.

Suitable materials which can be used as a substrate in the invention herein include, among others, sponges, paper, and woven and non-woven cloth, all having the necessary absorbency requirements defined above.

The preferred non-woven cloth substrates can generally be defined as adhesively bonded fibrous or filamentous products having a web or carded fiber structure (where the fiber strength is suitable to allow carding), or comprising fibrous mats in which the fibers or filaments are distributed haphazardly or in random array (i.e. an array of fibers is a carded web wherein partial orientation of the fibers is frequently present, as well as a completely

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haphazard distributional orientation), or substantially aligned. The fibers or filaments can be natural (e.g. wool, silk, jute, hemp, cotton, linen, sisal, or ramie) or synthetic (e.g. rayon, cellulose ester, polyvinyl derivatives, polyolefins, polyamides, or polyesters).

The preferred absorbent properties are particularly easy to obtain with non-woven cloths and are provided merely by building up the thickness of the cloth, i.e., by superimposing a plurality of carded webs or mats to a thickness adequate to obtain the necessary absorbent properties, or by allowing a sufficient thickness of the fibers to deposit on the screen. Any diameter or denier of the fiber (generally up to about 10 denier) can be used, inasmuch as it is the free space between each fiber that makes the thickness of the cloth directly related to the absorbent capacity of the cloth, and which, further, makes the non-woven cloth especially suitable for impregnation with a composition by means of intersectional or capillary action. Thus, any thickness necessary to obtain the required absorbent capacity can be used.

When the substrate for the composition is a non-woven cloth made from fibers deposited haphazardly or in random array on the screen, the articles exhibit excellent strength in all directions and are not prone to tear or separate when used in the automatic clothes dryer.

Preferably, the non-woven cloth is water-laid or air-laid and is made from cellulosic fibers, particularly from regenerated cellulose or rayon. Such non-woven cloth can be lubricated with any standard textile lubricant.

Preferably, the fibers are from 5mm to 50mm in length and are from 1.5 to 5 denier. Preferably, the fibers are at least partially orientated haphazardly, and are adhesively bonded together with a hydrophobic or substantially hydrophobic binder-resin. Preferably, the cloth comprises about 70% fiber and 30% binder resin polymer by weight and has a basis weight of from about 18 to 45g per square meter.

In applying the fabric softener composition to the absorbent substrate, the amount impregnated into and/or coated onto the absorbent substrate is conveniently in the weight ratio range of from about 10:1 to 0.5:1 based on the ratio of total conditioning composition to dry, untreated substrate (fiber plus binder). Preferably, the amount of the conditioning composition ranges from about 5:1 to about 1:1, most preferably from about 3:1 to 1:1, by

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weight of the dry untreated substrate.

According to one preferred embodiment of the invention, the dryer sheet substrate is coated by being passed over a rotogravure applicator roll. In its passage over this roll, the sheet is coated with a thin, uniform layer of molten fabric softening composition contained in a rectangular pan at a level of about 15g per square yard. Passage for the substrate over a cooling roll then solidifies the molten softening composition to a solid. This type of applicator is used to obtain a uniform homogeneous coating across the sheet.

Following application of the liquefied composition, the articles are held at room temperature until the composition substantially solidifies. The resulting dry articles, prepared at the composition substrate ratios set forth above, remain flexible; the sheet articles are suitable for packaging in rolls. The sheet articles can optionally be slitted or punched to provide a non-blocking aspect at any convenient time if desired during the manufacturing process.

The fabric softener composition employed in the present invention includes certain fabric softeners which can be used singly or in admixture with each other.

Examples of suitable textile fibre materials which can be treated with the fabric softener composition are materials made of silk, wool, polyamide, acrylics or polyurethanes, and, in particular, cellulosic fibre materials of all types. Such fibre materials are, for example, natural cellulose fibres, such as cotton, linen, jute and hemp, and regenerated cellulose. Preference is given to textile fibre materials made of cotton. The fabric softener compositions are also suitable for hydroxyl-containing fibres which are present in mixed fabrics, for example mixtures of cotton with polyester fibres or polyamide fibres.

A better understanding of the present invention and of its many advantages will be had by referring to the following Examples, given by way of illustration. The percentages given in the examples are percentages by weight.

## Example 1 (preparation of the rinse conditioners)

The liquid rinse conditioners are prepared by using the procedure described below. This type of fabric rinse conditioners is normally known under the name of "triple strength" or "triple fold" formula.

75 % by weight of the total amount of water is heated to 40°C. The molten fabric softener di-(palmcarboxyethyl-)hydroxyethyl-methylammonium-methosulfate (or Rewoquat WE 38 DPG available from Witco) is added to the heated water under stirring and the mixture is stirred for 1 hour at 40°C. Afterwards the aqueous softener solution is cooled down to below 30°C while stirring. When the solution cools down sufficiently magnesium chloride is added and the pH is adjusted to 3.2 with 0.1 N hydrochloric acid. The formulation is then filled up with water to 100%.

The rinse conditioner formulation as described above was used as a base formulation. In a final step the fabric softener is mixed with a separately prepared polyorganosiloxane /additive emulsion. The fabric softener formulations used in the following examples are listed in the following Table 1.

Table 1 (rinse conditioner formulations used in the application test for 1 kg wash load)

Rinse conditioner	Polyorgano-siloxane	Fabric softener	рН
formulation	emulsion (calculated	Base Formulation	
	on solid content of		
	the emulsion)		
0 (Reference)		13.3 g	3.2
Α	0.2 g of Type I	13.3 g	3.2
В	0.2 g of Type II	13.3 g	3.2
С	0.2 g of Type III	13.3 g	3.2
D	0.2 g of Type IV	13.3 g	3.2
E	0.2 g of Type V	13.3 g	3.2
F	0.2 g of Type VI	13.3 g	3.2
G	0.2 g of Type VII	13.3 g	3.2
Н	0.2 g of Type VIII	13.3 g	3.2
1	0.2 g of Type IX	13.3 g	3.2
J	0.2 g of Type X	13.3 g	3.2
K	0.2 g of Type XI	13.3 g	3.2
L	0.2 g of Type XII	13.3 g	3.2
М	0.2 g of Type XV	13.3 g	3.2

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## Types of polyorganosiloxane emulsions used

#### Type I

- Polyorganosiloxane of general formula (1), wherein R<sub>1</sub> is -OH, R<sub>3</sub> is -CH<sub>3</sub>,
- X + Y = 300-1500, % nitrogen (with respect to silicone) = 0
- 3.7% of an emulsifier
- 12.5% of an emulsifiable oxidised polyethylene which has a density of 0.95 to 1.05 g/cm<sup>3</sup> at 20°C, a drop point of 100-150°C, an acid number of 10 to 60 and a saponification number of 15 to 80
- solid content of the emulsion measured by evaporation at 120°C = 27.0-29.0%
- water content = 71.3%

#### Type II

- Polyorganosiloxane of general formula (1), wherein  $R_1$  is -OH,  $R_3$  is -CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>NH<sub>2</sub>, X + Y = 300-1500, % nitrogen (with respect to silicone) = 0.025
- 4.5% of an emulsifier
- 1% of an emulsifiable oxidised polyethylene which has a density of 0.95 to 1.05 g/cm<sup>3</sup> at 20°C, a drop point of 100-150°C, an acid number of 10 to 60 and a saponification number of 15 to 80
- solid content of the emulsion measured by evaporation at 120°C = 37.0-39.0%
- water content = 60.7%

#### Type III

- Polyorganosiloxane of general formula (1), wherein R<sub>1</sub> is -OH,

 $R_3$  is  $-CH_2CH_2CH_2N(H)(CH_2CH_2NH_2)$ , X + Y = 300-1500,

% nitrogen (with respect to silicone) = 0.03

- 3.6% of an emulsifier
- 14% of an emulsifiable oxidised polyethylene which has a density of 0.95 to 1.05 g/cm<sup>3</sup> at 20°C, a drop point of 100-150°C, an acid number of 10 to 60 and a saponification number of 15 to 80
- solid content of the emulsion measured by evaporation at 120°C = 23.0-25.0%
- water content = 73.7%

 $= \frac{4^{2} \left( \frac{1}{2} \left( \frac{1}{4} \right)^{2} \left( \frac{1}{4} \right)^{2} + \frac{1}{4^{2}} \left( \frac{1}{4} \right)^{2} + \frac{1}{4^{2}} \left( \frac{1}{4} \right)^{2} \left( \frac{1}{4} \right)^{$ 

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#### Type IV

- Polyorganosiloxane of general formula (1), wherein R<sub>1</sub> is -OH,

 $R_3$  is  $-CH_2CH_2CH_2N(H)(CH_2CH_2NH_2)$ , X + Y = 300-1500,

% nitrogen (with respect to silicone) = 0.11

- 4.3% of an emulsifier
- 0.3% of a fatty acid monoalkanolamide of formula (15b), wherein  $R_{34}$  is hydrogen and  $R_{37}$  is hydrogen or a radical of formula -C(O) $R_{36}$
- solid content of the emulsion measured by evaporation at 120°C = 37.0-39.0%
- water content = 60.7%

#### Type V

- Polyorganosiloxane of general formula (1), wherein R<sub>1</sub> is -OH,

 $R_3$  is  $-CH_2CH_2CH_2N(H)(CH_2CH_2NH_2)$ , X + Y = 300-1500,

% nitrogen (with respect to silicone) = 0.11

- 4.4% of an emulsifier
- 0.2% of an emulsifiable oxidised polyethylene which has a density of 0.95 to 1.05 g/cm<sup>3</sup> at 20°C, a drop point of 100-150°C, an acid number of 10 to 60 and a saponification number of 15 to 80
- solid content of the emulsion measured by evaporation at 120°C = 37.0-39.0%
- water content = 60.7%

#### Type VI

- Polyorganosiloxane of general formula (1), wherein R<sub>1</sub> is -CH<sub>3</sub>,

 $R_3$  is  $-CH_2CH_2CH_2N(H)(CH_2CH_2NH_2)$ , X + Y = 150-300,

% nitrogen (with respect to silicone) = 0.12

- 11% of an emulsifier
- 0.3% of a fatty acid dialkanolamide of formula (15a), wherein  $R_{34}$ ,  $R_{38}$ ,  $R_{38}$  and  $R_{38}$  are hydrogen or -CH<sub>2</sub>OH
- solid content of the emulsion measured by evaporation at 120°C = 24.0-26.0%
- water content = 72.1%

#### Type VII

- Polyorganosiloxane of general formula (1), wherein R<sub>1</sub> is -CH<sub>3</sub>,

 $R_3$  is  $-CH_2CH_2CH_2N(H)(CH_2CH_2NH_2)$ , X + Y = 40-150,

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% nitrogen (with respect to silicone) = 0.08

- 13.2% of an emulsifier
- 0.23% of an emulsifiable oxidised polyethylene which has a density of 0.95 to 1.05 g/cm<sup>3</sup> at 20°C, a drop point of 100-150°C, an acid number of 10 to 60 and a saponification number of 15 to 80
- solid content of the emulsion measured by evaporation at 120°C = 41.0-43.0%
- water content = 44.4%

#### Type VIII

- Polyorganosiloxane of general formula (1), wherein  $R_1$  is -CH<sub>3</sub>,  $R_3$  is -CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>N(H)(CH<sub>2</sub>CH<sub>2</sub>N(H)(CO)(CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>OH))), X + Y = 300-1500, % nitrogen (with respect to silicone) = 0.1
- 9.8% of an emulsifier
- 0.1% of an emulsifiable oxidised polyethylene which has a density of 0.95 to 1.05 g/cm<sup>3</sup> at 20°C, a drop point of 100-150°C, an acid number of 10 to 60 and a saponification number of 15 to 80
- solid content of the emulsion measured by evaporation at 120°C = 20.5-22.5%
- water content = 76.9%

#### Type IX

- Polyorganosiloxane of general formula (8), wherein  $R_{17}$  is -CH<sub>3</sub>,  $R_3$  is CH<sub>3</sub>,  $R_{19}$  is a polyethylenoxide radical,  $X^1 + Y^1 + S = 40-150$ , % nitrogen (with respect to silicone) = 0
- 2% of an emulsifier
- 0.15% of an emulsifiable oxidised polyethylene which has a density of 0.95 to 1.05 g/cm<sup>3</sup> at 20°C, a drop point of 100-150°C, an acid number of 10 to 60 and a saponification number of 15 to 80
- solid content of the emulsion measured by evaporation at 120°C = 23.0-25.0%
- water content = 74.9%

#### Type X

- Polyorganosiloxane of general formula (8), wherein  $R_{17}$  is -CH<sub>3</sub>,  $R_3$  is -CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>NH<sub>2</sub>,  $R_{19}$  is a polyethylene/polypropyleneoxide radical,  $X^1 + Y^1 + S = 150-300$ 

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% nitrogen (with respect to silicone) = 0.07

- 3.5% of an emulsifier
- 1.5% of a fatty acid dialkanolamide of formula (15a), wherein  $R_{34}$ ,  $R_{38}$ ,  $R_{38}$ ' and  $R_{38}$ " are hydrogen or -CH<sub>2</sub>OH
- solid content of the emulsion measured by evaporation at 120°C = 19.5-21.5%
- water content = 73%

#### Type XI

- Polyorganosiloxane of general formula (9), wherein  $R_{26}$  is  $C_{12}$ alkyl,  $R_{27}$  is 2-phenylpropyl,  $R_{28}$  is an epoxy radical of formula (10),  $X^2 + X^3 + X^4 + Y^2 = 40$ -150, % nitrogen (with respect to silicone) = 0
- 2.9% of an emulsifier
- 0.85% of an emulsifiable oxidised polyethylene which has a density of 0.95 to 1.05 g/cm<sup>3</sup> at 20°C, a drop point of 100-150°C, an acid number of 10 to 60 and a saponification number of 15 to 80
- solid content of the emulsion measured by evaporation at 120°C = 37.0-39.0%
- water content = 62%

#### Type XII

- Polyorganosiloxane of general formula (1), wherein R<sub>1</sub> is CH<sub>3</sub>, R<sub>3</sub> is C<sub>18</sub>alkoxy,
- X + Y = 40-150, % nitrogen (with respect to silicone) = 0
- 3.2% of an emulsifier
- 1.5% of an emulsifiable oxidised polyethylene which has a density of 0.95 to 1.05 g/cm<sup>3</sup> at 20°C, a drop point of 100-150°C, an acid number of 10 to 60 and a saponification number of 15 to 80
- solid content of the emulsion measured by evaporation at 120°C = 34.0-35.5%
- water content = 61.4%

#### Type XIII

- Polyorganosiloxane of general formula (1), wherein R<sub>1</sub> is -OH,
- $R_3$  is  $-CH_2CH_2CH_2N(H)(CH_2CH_2NH_2)$ , X + Y = 300-1500,
- % nitrogen (with respect to silicone) = 0.1
- 4.2% of an emulsifier

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- 6.2% of a fatty acid monoalkanolamide of formula (15b), wherein  $R_{34}$  is hydrogen and  $R_{37}$  is hydrogen or a radical of formula -C(O) $R_{36}$
- solid content of the emulsion measured by evaporation at 120°C = 38-40%
- water content = 60%

#### Type XIV

- Polyorganosiloxane of general formula (8), wherein  $R_3$  is -CH<sub>3</sub> and  $R_{17}$  is -CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>NH<sub>2</sub>,  $R_{19}$  is a polyethyleneoxide radical,  $X^1 + Y^1 + S = 40-150$ ,
- % nitrogen (with respect to silicone) = 0.04
- 7.2% of an emulsifier
- solid content of the emulsion measured by evaporation at 120°C = 54-56%
- water content = 38.1%

#### Type XV

1 part of emulsion Type XIII and 9 parts of emulsion type XIV

#### Example 2 Hydrophilicity

The formulated rinse conditioners (see Table 1) are applied according to the following procedure:

Woven cotton swatches of size of 50 cm by 40 cm are washed together with ballast material (cotton and cotton/polyester) in a AEG Oeko Lavamat 73729 washing machine maintaining the washing temperature at 40°C. The total fabric load of 1 kg is washed for 15 minutes with 33 g of ECE Color Fastness Test Detergent 77 (Formulation January 1977, according to ISO 105-CO6). The rinse conditioner formulation as described in Table 1 is applied in the last rinse cycle at 20°C. After rinsing with the formulation the textile swatches are dried on a washing line at ambient temperature.

#### **Evaluation of Hydrophilicity**

The water absorption of fabrics treated with the test samples is measured by the wicking test. Test strips are fixed to a frame and dipped about 1mm deep in a colored aqueous solution. The rise of water in the strips is measured after twenty minutes. Water absorption

of fabrics treated with rinse conditioner formulations from Table 1 are compared. The average values of four parallel measurements are given in Table 2.

Table 2

Rinse conditioner sample	Measured water rise				
	(cm)				
0 (Reference)	8.2				
Α	9.6				
В	10.2				
С	9.5				
D	9.6				
E	9.2				
F	9.0				
G	9.8				
Н	9.1				
ı	9.2				
J	9.6				
К	9.8				
L	9.6				
M	9.3				

These results show an improved hydrophilicity of the textile fabric material treated with compositions of the present invention.

In all experiments the following textiles have been used:

Cotton woven: 120 g/m2, bleached, with resin finishing:

Cotton/Polyester 66/34 woven: 85 g/m2, bleached.

Both textiles were finished with a resin according to Oekotex Standard 100: 30 g/l of modified dimethyloldihydroxyethylene urea (70% active material) 9 g/l Magnesiumchloride (with 6 H<sub>2</sub>O) padding with a pick-up of approximately 80%

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Drying at about 110 - 120 °C in a oven followed by a 4 minute curing step at 145°C.

#### WHAT IS CLAIMED IS:

- 1. A method of use of a softener composition for imparting hydrophilicity to textile fibre materials in domestic applications, which softener composition comprises:
- A) a fabric softener;
- B) at least one additive selected from the group consisting of
  - a) a polyethylene, or a mixture thereof,
  - b) a fatty acid alkanolamide, or a mixture thereof,
  - c) a polysilicic acid, or a mixture thereof, and
  - d) a polyurethane, or a mixture thereof; and
- C) a dispersed polyorganosiloxane of formula (1)

(1) 
$$R^{1} - S_{i} - O = \begin{bmatrix} CH_{3} \\ I \\ S_{i} - O \end{bmatrix} = \begin{bmatrix} CH_{3} \\ I \\ S_{i} - O \end{bmatrix} = \begin{bmatrix} CH_{3} \\ I \\ S_{i} - O \end{bmatrix} = \begin{bmatrix} CH_{3} \\ I \\ S_{i} - O \end{bmatrix} = \begin{bmatrix} CH_{3} \\ I \\ CH_{3} \end{bmatrix}$$

#### wherein

R1 is OH, OR2 or CH3

R2 is CH3 or CH2CH3

R<sup>3</sup> is C<sub>1</sub>-C<sub>20</sub>alkoxy, CH<sub>3</sub>, CH<sub>2</sub>CHR<sup>4</sup>CH<sub>2</sub>NHR<sup>5</sup>, or CH<sub>2</sub>CHR<sup>4</sup>CH<sub>2</sub>N(COCH<sub>3</sub>)R<sup>5</sup>

(2) 
$$(CH_2)_3O$$
 NR<sup>8</sup>

or (3)  $(CH_2)_3NH$  CH

or (4)  $(CH_2)_3$  NR<sup>8</sup>

R4 is H or CH3

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 $R^5$  is H,  $CH_2CH_2NHR^6$ ,  $C(=O)-R^7$  or  $(CH_2)_Z$ - $CH_3$  z is 0 to 7  $R^6$  is H or  $C(=O)-R^7$   $R^7$  is  $CH_3$ ,  $CH_2CH_3$  or  $CH_2CH_2CH_2OH$   $R^8$  is H or  $CH_3$  the sum of X and Y is 40 to 4000;

or a dispersed polyorganosiloxane which comprises at least one unit of the formula (5)

(5) 
$$(R^9)_V (R^{10})_W \text{Si-A-B}$$

wherein

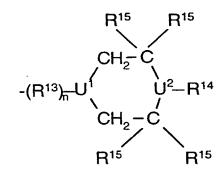
R9 is CH3, CH3CH2 or Phenyl

R<sup>10</sup> is -O-Si or -O-R<sup>9</sup>

the sum of v and w equals 3, and v does not equal 3

 $A = -CH_2CH(R^{11})(CH_2)_K$ 

 $B = -NR^{12}((CH_2)_l-NH)_mR^{12}$ , or



n is 0 or 1

(6)

when n is 0, U1 is N, when n is 1, U1 is CH

l is 2 to 8

k is 0 to 6

m is 0 to 3

R<sup>11</sup> is H or CH<sub>3</sub>

 $R^{12}$  is H, C(=O)- $R^{16}$ , CH<sub>2</sub>(CH<sub>2</sub>)<sub>p</sub>CH<sub>3</sub> or

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p is 0 to 6

R<sup>13</sup> is NH, O, OCH<sub>2</sub>CH(OH)CH<sub>2</sub>N(Butyl), OOCN(Butyl)

R14 is H, linear or branched C1-C4 alkyl, Phenyl or CH2CH(OH)CH3

R15 is H or linear or branched C1-C4 alkyl

R<sup>16</sup> is CH<sub>3</sub>, CH<sub>2</sub>CH<sub>3</sub> or (CH<sub>2</sub>)<sub>q</sub>OH

q is 1 to 6

U<sup>2</sup> is N or CH:

or a dispersed polyorganosiloxane of the formula (8)

(8) 
$$R^{17} - Si - O = \begin{bmatrix} CH_3 \\ | \\ | \\ CH_3 \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ | \\ CH_3 \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix} CH_3 \\ | \\ Si - O \end{bmatrix} = \begin{bmatrix}$$

#### wherein

R<sup>3</sup> is as previously defined

R<sup>17</sup> is OH, OR<sup>18</sup> or CH<sub>3</sub>

R<sup>18</sup> is CH<sub>3</sub> or CH<sub>2</sub>CH<sub>3</sub>

 $R^{19}$  is  $R^{20}$ -(EO)<sub>m</sub>-(PO)<sub>n</sub>- $R^{21}$ 

m is 3 to 25

n is 0 to 10

R<sup>20</sup> is the direct bond or CH<sub>2</sub>CH(R<sup>22</sup>)(CH<sub>2</sub>)<sub>0</sub>R<sup>23</sup>

p is 1 to 4

R<sup>21</sup> is H, R<sup>24</sup>, CH<sub>2</sub>CH(R<sup>22</sup>)NH<sub>2</sub> or CH(R<sup>22</sup>)CH<sub>2</sub>NH<sub>2</sub>

R<sup>22</sup> is H or CH<sub>3</sub>

R<sup>23</sup> is O or NH

R<sup>24</sup> is linear or branched C<sub>1</sub>-C<sub>8</sub> alkyl or Si(R<sup>25</sup>)<sub>3</sub>

R<sup>25</sup> is R<sup>24</sup>, OCH<sub>3</sub> or OCH<sub>2</sub>CH<sub>3</sub>

EO is -CH2CH2O-

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PO is  $-CH(CH_3)CH_2O$ - or  $-CH_2CH(CH_3)O$ the sum of  $X_1, Y_1$  and S is 20 to 1500;

or a dispersed polyorganosiloxane of the formula (9)

#### wherein

R<sup>26</sup> is linear or branched C<sub>1</sub> - C<sub>20</sub> alkoxy, CH<sub>2</sub>CH(R<sup>4</sup>)R<sup>29</sup>

R<sup>4</sup> is as previously defined

R<sup>29</sup> is linear or branched C<sub>1</sub> - C<sub>20</sub> alkyl

 $R^{27}$  is aryl, aryl substituted by linear or branched  $C_1$  -  $C_{10}$  alkyl, linear or branched  $C_1$  -  $C_{20}$  alkyl substituted by aryl or aryl substituted by linear or branched  $C_1$  -  $C_{10}$  alkyl  $R^{28}$  is

the sum of  $X^2$ ,  $X^3$ ,  $X^4$  and  $Y^2$  is 20 to 1500, wherein  $X^3$ ,  $X^4$  and  $Y^2$  may be independently of each other 0;

or a mixture thereof.

## 2. A method of use according to claim 1 wherein the polyorganosiloxane is of formula (1):

(1) 
$$R^{1} - S_{1} - O = \begin{bmatrix} CH_{3} \\ | \\ S_{1} - O \end{bmatrix} = \begin{bmatrix} CH_{3} \\ | \\ S_{1} - O \end{bmatrix} = \begin{bmatrix} CH_{3} \\ | \\ S_{1} - O \end{bmatrix} = \begin{bmatrix} CH_{3} \\ | \\ S_{1} - O \end{bmatrix} = \begin{bmatrix} CH_{3} \\ | \\ CH_{3} \end{bmatrix} = \begin{bmatrix} CH_{$$

#### wherein

R<sup>1</sup> is OH, OR<sup>2</sup> or CH<sub>3</sub>

R<sup>2</sup> is CH<sub>3</sub> or CH<sub>2</sub>CH<sub>3</sub>

R<sup>3</sup> is C<sub>1</sub>-C<sub>20</sub>alkoxy, CH<sub>3</sub>, CH<sub>2</sub>CHR<sup>4</sup>CH<sub>2</sub>NHR<sup>5</sup>, or

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(2) 
$$(CH_2)_3O$$
 NR<sup>8</sup>

Or (3)  $(CH_2)_3NH$  CH

 $R^4$  is H or CH<sub>3</sub>  $R^5$  is H, CH<sub>2</sub>CH<sub>2</sub>NHR<sup>6</sup>, C(=O)-R<sup>7</sup>  $R^6$  is H or C(=O)-R<sup>7</sup>  $R^7$  is CH<sub>3</sub>, CH<sub>2</sub>CH<sub>3</sub> or CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>OH  $R^8$  is H or CH<sub>3</sub> the sum of X and Y is 40 to 4000;

or a dispersed polyorganosiloxane which comprises at least one unit of the formula (5);

(5) 
$$(R^9)_V (R^{10})_W \text{Si-A-B}$$

wherein

(6)

 $R^9$  is  $CH_3$ ,  $CH_3CH_2$   $R^{10}$  is -O-Si or -O-R<sup>9</sup> the sum of v and w equals 3, and v does not equal 3  $A = -CH_2CH(R^{11})(CH_2)_K$ B =

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n is 1 U<sup>1</sup> is CH

k is 0 to 6

R<sup>11</sup> is H or CH<sub>3</sub>

R<sup>13</sup> is OOCN(Butyl)

R<sup>14</sup> is H, linear C<sub>1</sub>-C<sub>4</sub> alkyl, Phenyl

R15 is H or linear C1-C4 alkyl

U<sup>2</sup> is N;

or a dispersed polyorganosiloxane of the formula (8);

(8) 
$$R^{17} - Si - O = CH_3$$
  $CH_3$   $CH_4$   $CH_3$   $CH_4$   $CH_4$   $CH_5$   $CH_5$ 

#### wherein

R<sup>3</sup> is as previously defined

R<sup>17</sup> is OH, OR<sup>18</sup> or CH<sub>3</sub>

R<sup>18</sup> is CH<sub>3</sub> or CH<sub>2</sub>CH<sub>3</sub>

 $R^{19}$  is  $R^{20}$ -(EO)<sub>m</sub>-(PO)<sub>o</sub>- $R^{21}$ 

m is 3 to 25

n is 0 to 10

R<sup>20</sup> is the direct bond or CH<sub>2</sub>CH(R<sup>22</sup>)(CH<sub>2</sub>)<sub>p</sub>R<sup>23</sup>

p is 1 to 4

R<sup>21</sup> is H, R<sup>24</sup>, CH<sub>2</sub>CH(R<sup>22</sup>)NH<sub>2</sub> or CH(R<sup>22</sup>)CH<sub>2</sub>NH<sub>2</sub>

R<sup>22</sup> is H or CH<sub>3</sub>

R<sup>23</sup> is O or NH

R<sup>24</sup> is linear or branched C<sub>1</sub>-C<sub>3</sub> alkyl or Si(R<sup>25</sup>)<sub>3</sub>

R<sup>25</sup> is R<sup>24</sup>, OCH<sub>3</sub> or OCH<sub>2</sub>CH<sub>3</sub>

EO is -CH2CH2O-

PO is -CH(CH<sub>3</sub>)CH<sub>2</sub>O- or -CH<sub>2</sub>CH(CH<sub>3</sub>)O-

the sum of  $X_1, Y_1$  and s is 20 to 1500;

or a dispersed polyorganosiloxane of the formula (9);

#### wherein

R<sup>26</sup> is linear C<sub>1</sub> - C<sub>20</sub> alkoxy,

R4 is as previously defined

R<sup>29</sup> is linear C<sub>1</sub> - C<sub>20</sub> alkyl

R<sup>27</sup> is, CH<sub>2</sub>CH(R<sup>4</sup>)Phenyl

R<sup>28</sup> is

(10) 
$$(CH_2)_3$$
  $-CH_2$   $-CH_2$   $-CH_2$   $-CH_2$ 

the sum of  $X^2$ ,  $X^3$ ,  $X^4$  and  $Y^2$  is 20 to 1500, wherein  $X^3$ ,  $X^4$  and  $Y^2$  may be independently of each other 0;

or a mixture thereof.

3. A method of use according to claim 1 or 2 wherein a polyorganosiloxane of formula (1) is used, wherein

R1 is OH or CH3,

R<sup>3</sup> is CH<sub>3</sub>, C<sub>10</sub>-C<sub>20</sub>alkoxy or CH<sub>2</sub>CHR<sup>4</sup>CH<sub>2</sub>NHR<sup>5</sup>,

R⁴ is H.

R<sup>5</sup> is H or CH<sub>2</sub>CH<sub>2</sub>NHR<sup>6</sup>,

R<sup>6</sup> is H or C(=0)-R<sup>7</sup>, and

R<sup>7</sup> is CH<sub>3</sub>, CH<sub>2</sub>CH<sub>3</sub> or especially CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>OH.

4. A method of use according to claim 1 or 2 wherein a polyorganosiloxane of formula (8) is used, wherein

R<sup>3</sup> is CH<sub>3</sub>, C<sub>10</sub>-C<sub>20</sub>alkoxy or CH<sub>2</sub>CHR<sup>4</sup>CH<sub>2</sub>NHR<sup>5</sup>,

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 $R^4$  is H,  $R^5$  is H or  $CH_2CH_2NHR^6$ ,  $R^6$  is H or  $C(=O)-R^7$ ,  $R^7$  is  $CH_2CH_3$ ,  $CH_2CH_2CH_2OH$  or especially  $CH_3$ , and  $R_{17}$  is  $CH_3$  or OH.

5. A method of use according to claim 1 or 2 wherein a polyorganosiloxane of formula (9) is used, wherein

R<sup>26</sup> is CH<sub>2</sub>CH(R<sup>4</sup>)R<sup>29</sup>,

R⁴ is H, and

R<sup>27</sup> is 2-phenyl propyl.

- 6. A method of use according to any of claims 1 to 5 wherein the composition is a liquid aqueous composition.
- 7. A method of use according to any of claims 1 to 5 wherein the composition is used in a tumble dryer sheet composition.
- 8. A method of use according to any of claims 1 to 7 in which the polyorganosiloxane is nonionic or cationic.
- 9. A method of use according to any of claims 1 to 8 in which the composition has a solids content of 5 to 70 % at a temperature of 120°C.
- 10. A method of use according to any of claims 1 to 9 in which the composition contains a water content of 25 to 90 % by weight based on the total weight of the composition.
- 11. A method of use according to any of claims 1 to 10 in which the composition has a pH value from 2 to 7.
- 12. A method of use according to any of claims 1 to 11 in which the nitrogen content of the aqueous emulsion due to the polyorganosiloxane is from 0 to 0.25 % with respect to the silicon content.

- 13. A method of use according to any of claims 1 to 12 wherein the composition comprises a polyethylene, a fatty acid alkanolamide or a polyurethane.
- 14. A method of use according to any of claims 1 to 13 wherein the composition comprises a polyethylene or a fatty acid alkanolamide.
- 15. A method of use according to any of claims 1 to 14 wherein the composition comprises a fatty acid alkanolamide.
- 16. A method of use according to any of claims 1 to 14 wherein the composition comprises a polyethylene.
- 17. A method of use according to any of claims 1 to 16 wherein the composition is prepared by mixing a preformulated fabric softener with an emulsion comprising the polyorganosiloxane and the additive.
- 18. A method of use according to any of claims 1 to 17 wherein composition has a clear appearance.
- 19. A method of use according to any of claims 1 to 18 in which the composition comprises:
- a) 0.01 to 70 % by weight, based on the total weight of the composition, of a polyorganosiloxane, or a mixture thereof;
- b) 0.2 to 15 % by weight based on the total weight of an emulsifier, or a mixture thereof;
- c) 0.01 to 15 % by weight based on the total weight of at least one additive selected from the group consisting of a polyethylene, a fatty acid alkanolamide, a polysilicic acid and a polyurethane, and
- d) water to 100 %.
- 20. A tumble dryer sheet comprising a composition as defined in claim 1.





## (12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

# (19) World Intellectual Property Organization International Bureau

# AIPO OMPI

## 

## (43) International Publication Date 12 April 2001 (12.04.2001)

#### **PCT**

# (10) International Publication Number WO 01/25383 A1

(51) International Patent Classification<sup>7</sup>: 17/04, 3/12, 1/645, 1/62

C11D 3/37,

(21) International Application Number: PCT/EP00/09396

(22) International Filing Date:

26 September 2000 (26.09.2000)

(25) Filing Language:

English

(26) Publication Language:

English

(30) Priority Data: 99810899.7

5 October 1999 (05.10.1999) EP

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- (81) Designated States (national): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CR, CU, CZ, DE, DK, DM, DZ, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.
- (84) Designated States (regional): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

#### Published:

With international search report.

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

11/25383 A1

(54) Title: FABRIC SOFTENER COMPOSITIONS

(57) Abstract: The present invention relates to a method of use of a softener composition for imparting hydrophylicity to textile fibre materials in domestic applications, which softener composition comprises: A) a fabric softener; B) at least one additive selected from the group consisting of a) a polyethylene, or a mixture thereof, b) a fatty acid alkanolamide, or a mixture thereof, c) a polysilicic acid, and d) a polyurethane; C) selected polyorganosiloxanes.



US Case HF/5 -22102/US/A

#### **DECLARATION AND POWER OF ATTORNEY FOR U.S. PATENT APPLICATIONS**

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As a	As a below named inventor, I hereby declare that:								
My r	esiden	ce, post off	ice address a	nd citizensh	nip a	re as st	ated below ne	xt to	my name.
I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if more than one name is listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled									
FAB	RIC SO	FTENER CO	MPOSITIONS						
whic	h is de	scribed and	l claimed in:						
	the attached specification.								
	the s filed	specification (day/mon		ication No. d as amend	led (	on _	(day/month/yea		if applicable).
×	the s filed	specification 26/09 (day/mont		nal Applicat	tion	No.	PCT/EP 00/09	396	
	assi	gned U.S. A	Application No	).			(if applicable)	, and	d as amended
		under PCT	Article 19 on	(day/m	onth	/year)	(if applicable)	)	
		under PCT	Article 34 on	(day/m	onth	/year)	(if applicable)	)	
		and further	amended on	(day/m	onth	/year)	(if applicable)	ł	

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose all information which is known by me to be material to the patentability of this application as defined in 37 C.F.R. § 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. § 119 (a)-(d) of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America relating to this subject matter having a filing date before that of the application on which priority is claimed:

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COUNTRY/REGION (OR PCT)	APPLICATION No		FILING (day/mor			PRI	ORIT	Y CLA	IMED
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							Yes		No
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I hereby claim the ben- application(s) listed be		§ 1	19 (e) of an	y Un	ited Sta	ates	provis	sional	
APPLICATION NO.			FILING DA						
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PCT APPLICATION No. (designating the U.S.)	INTERNATIONAL FILING DATE (day/month/year)	L	U.S. APPLICATION STATUS No. (if any)						
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I hereby appoint the following attorneys and agents, associated with Customer No. 000324, each of them with full power of substitution, revocation and appointment of associates, to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. § 1001, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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